#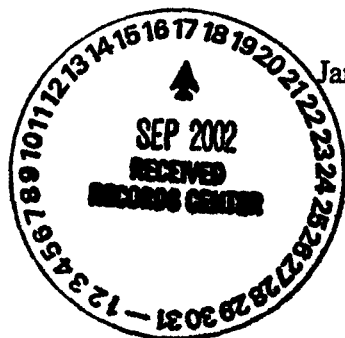 Soil Aggregation and Its Influence on ^{239,240}Pu Particle-Size Distributions of Soils Collected From Rocky Flats, CO

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Soil Aggregation and ^{239,240}Pu Particle-Size Distribution in Rocky Fats, CO, Soils

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1   EXECUTIVE SUMMARY

This report describes the results of an investigation into the role of soil aggregation in establishing the particle-size distribution of ^{239,240}Pu in Rocky Flats buffer-zone soils. The report also contains an analysis of particle aggregation in a single runoff sample   The following general conclusions were derived from the study.

1   *Between 60 and 70% of soil $^{239\,240}$Pu inventory (i.e., ΣPu) resides in the sand (> 53 \mu m)*
   fraction of undisturbed Rocky Flats soils(Figure 9) These aggregates are likely a consequence of the biological activity that occurs in an normal grassland soil. These aggregates are stable ' with respect to disaggregation upon emersion in water

2. *Results of the sonication analyses show that the water-stable aggregates can be mechanically dispersed.* The response of soil aggregates to sonication is an indication of the mechanical stability of the soils Sonication causes a substantial increase in the fraction of the $^{239\,240}$Pu inventory in the less than 10 \mu m fractions (Figure 9)   Mechanical stresses to Rocky Flats soils (e g., machinery operations, freeze-thaw cycles, desiccation and raindrop impacts) may result in changes to the size distribution of ^{239,240}Pu in the buffer zone soils

3   *The destruction of organic matter substantially shifts the distribution of $^{239\,240}$Pu to the respirable size fraction (Figure 23)* Hydrogen peroxide (H_2O_2) oxidation caused an even more substantial shift in the Pu size distribution than did sonication. After partial destruction of the soil organic matter by chemical oxidation, the percentage of $^{239\,240}$Pu in the less than 10 \mu m fraction increased to 75 % of the ^{239,240}Pu soil inventory This has significant implications for the issue of grassland fires at the Rocky Flats site if combustion causes soil organic matter oxidation, then the particle size association may shift to smaller particle sizes (i e , those more subject to transport and inhalation).

4. *The Pu distribution among the various size fractions of the runoff sample is substantially different from that found for the case of the water-dispersion of the soils (Figure 36).* In the runoff sample, $^{239\,240}$Pu was primarily present in the smallest size fractions Comparison of the soil and runoff results suggest that it is the < 2 \mu m fraction of soil that has the greatest potential impact on Pu transport The amount of $^{239\,240}$Pu in the colloid (2 \mu m to 10K Dalton) and dissolved (less than 10K Dalton) fractions, in the runoff sample, was roughly equal

5. *Finally, a strong correlation between $^{239\,240}$Pu activity and particle specific surface area does not exist (Figure 28).* This result indicates that Pu distribution in Rocky Flats soils is not a particle surface-controlled phenomena and brings to question the appropriateness of 'K_d' models of Pu association with soil constituents

tube rotator (Scientific Equipment Products) which turned the samples end-over-end for 4-5 hours. In the soil aggregates study, 50-100 grams of dry soil were placed into a 1 liter glass Erlenmeyer flask and 400 ml of dispersant solution was added. The sample was then agitated for 24 hours on an orbital shaker table (Thermolyne RotoMax type 50800). For the runoff study, dry dispersant chemicals were added directly to the samples in order to prevent dilution of the sample. The samples were then agitated for 24 hours on the orbital shaker table.

Deionized water. Addition of deionized water only allowed examination of the water stable aggregates present in the soil. In this case the aggregates that form during drying are partially destroyed by the wetting of the sample. This is the gentlest means of soil dispersion and represents the natural condition of the soil after wetting by precipitation or snow-melt. For the runoff sample, nothing was done to the sample except to place it on the shaker table.

Ultrasonication. In a second dispersion experiment, following the addition of deionized water, we used ultrasonication to examine the mechanical stability of the aggregates. Ultrasonication provides mechanical energy that breaks apart the aggregates but does not fracture primary particles. This is not the case with other mechanical means of disaggregation, such as grinding. For the watershed soils study, samples were sonicated using an ultrasonic cleaning bath (Branson, 125 Watts). The samples were sonicated for six ten-minute intervals spaced over the course of five hours. The samples were shaken overnight and sonicated for an additional 10 minutes just prior to size fractionation. For the soil aggregates and runoff studies the sonication procedure employed an ultrasonic probe (Fisher Scientific Sonic Dismembrator, 50 Watts), in addition to the bath in order to provide a greater amount of aggregate dispersion. The probe was used at a 40% output setting. Although the rated power of the probe is less than the bath, a greater amount of energy is transferred to the sample because the probe is immersed directly in the sample. In this procedure after the samples had been sonicated using the bath, they were sonicated with the probe for an additional 15 minutes. This was done using three 5 minute intervals spaced a minimum of 10 minutes apart. This procedure was followed to minimize heating of the sample.

that was used in the soil aggregates study, the hydrogen peroxide that was initially added was consumed before complete oxidation of the organic matter This was evidenced by the cessation of carbon dioxide formation and by the residual brown color of the sample Therefore additional hydrogen peroxide was added to return the solution to a computed hydrogen peroxide concentration of 2 5 % A third addition of hydrogen peroxide was performed after the carbon dioxide evolution again ceased At the end of the hydrogen peroxide dispersion some small amount of color remained in the solution suggesting an incomplete oxidation of the organic matter in this experiment For both the watershed and runoff studies, no observable color remained after the addition of the initial hydrogen peroxide The pH of the slurries in the soil aggregates and runoff studies was held at near neutrality by the addition of sufficient sodium carbonate to obtain a computed solution-phase concentration of 0 01 M The final pH of the solutions were 7 2 and 6 5 for the soil and runoff samples, respectively

A final disaggregation experiment was performed that was designed to remove any iron oxide cements that may be holding the soil aggregates together This procedure can also release organic matter into solution, especially the fraction that is associated with iron oxides In this experiment the samples were treated with a combination of sodium citrate, sodium dithionite, and sodium bicarbonate The procedure was a modification of the method outlined by Gee and Bauder (1986) For the watershed soil samples the final concentrations of CDB were 0 15, 0 05, and 0 1 M, respectively For the soil aggregates study the citrate concentration used was reduced to 0 1 M The concentrations of all the reagents were reduced for the runoff sample because of the small amounts of suspended matter present Concentrations of CDB used for the runoff sample were 0 01, 0 006, and 0 003 M respectively

2 4 Size Fractionation Methods

The three methods of size fractionation used were wet sieving, sedimentation, and tangential flow filtration (TFF) Stainless steel sieves (8 inch diameter) were used to separate the samples into fractions having size ranges of 2000-212 (coarse sand), 212-53 (fine sand), and 53-25 (coarse silt) micrometers The sieves were stacked with the largest mesh size on top of the stack The

about 1 micron to settle The supernatant liquid was removed and the settled material transferred to the cylinder This allowed use of the supernatant liquid instead of deionized water for the triplicate settling experiments and therefore maintained the volume at about 2 liters

The final size fractionation involved the use of a Millipore MiniTan II tangential flow filtration system Two pores sizes, 0 45 micrometer and 10,000 Dalton MW, were used to separate the samples into coarse colloid (2-0 45 micrometer), fine colloid (0 45 um -10 K Dalton), and "dissolved" (< 10K Dalton) The filter membranes were soaked over night in deionized water and then were further washed by filtering 2-3 liters of deionized water prior to use Tangential flow filtration was performed by recirculating the sample over the surface of the filter membrane in order to maintain a clean filter surface By slightly constricting the return flow, a backpressure was created that forced some filtrate through the membrane The ratio of recirculation and filtrate flow rates was about 10 to 1 Filtration was performed until the retentate volume (sample which did not pass through the filter) was reduced to about 100-150 ml The filter membrane was then removed and placed in a Teflon bag along with a small amount of filtrate By rubbing the filter from outside the bag the soil that was coating the membrane was removed This material was then transferred to the retentate container

All of the various suspensions obtained from the size fractionation procedure were dried on a hotplate The volume of suspension and the mass of each dried fraction was obtained

2 5 Size Analysis (Single Particle Optical Sensing)

Particle size analysis was performed on selected samples using a single particle optical sensor (SPOS) This instrument is used to determine the size of each particle in a dilute sample based on the change in the detector signal as a particle passes between a laser light source and a series of two photodiodes The detector response is a result of a combination of light blockage and light scattering Small aliquots of the sample, about 10-100 microliter, were diluted to 1 liter The SPOS instrument then draws the sample through a capillary where the particles are detected By use of a high-speed pulse counter, the individual counts are summed and a size distribution is obtained The SPOS method was used in the watershed study to estimate the amount of < 2

2 8 BET Surface Area Analysis

Specific surface area of soil size fractions was determined by single point BET surface area measurement using a Micrometrics Flowsorb II 2300 BET surface area analyzer Analytical protocols conform to ASTM Method D-4567-86, "Standard test method for single-point determination of specific surface area of catalysts using nitrogen adsorption by continuous flow method" (ASTM, 1994) In this method, the solid sample is degassed by heating in a flow of He/N_2 The sample is then immersed in a liquid nitrogen bath causing adsorption of nitrogen from a flowing mixture of a fixed concentration of nitrogen in helium When adsorption is complete, the sample is allowed to warm to room temperature causing desorption The quantity of nitrogen gas desorbed is determined by sensing the change in thermal conductivity Specific surface area of the sample is calculated based on a modified form of the BET equation

2.9 Metals analysis.

Fractions of the actinide-containing solutions were taken from the digested samples for metals analyses The fractions were diluted with 1 M nitric acid to approximately 15 mL and then submitted for direct aspiration and quantification by inductively coupled plasma emission spectrometry All metals analyses under this study were performed on a Perkin Elmer Optima 3000 inductively coupled plasma-emission spectrometer with a Perkin Elmer AS 91 Auto Sampler. The system software provides two techniques for minimizing spectral interferences inter-element correction and multi-component spectral fitting Metals were analyzed per Perkin Elmer specifications using standard protocols Quality Assurance measures for these analyses include initial calibration with NIST traceable standards, continuing calibration verification throughout the analytical run time Scandium is utilized as an internal spike for assessing performance parameters Data review was performed by qualified ICP operators and the ICP laboratory supervisor prior to final reporting

The percent mass of the < 2 μm fraction was calculated from the volume fraction of particles in the SPOS-generated size distributions for each soil   By assuming a uniform particle density, the mass distribution can be considered equivalent to the volume distribution   The percent mass distribution results for the deionized water, sonication, and hydrogen peroxide analyses are shown in Figure 6   Data for the upper-most and lower-most five watershed samples are combined in Table 1

As can be seen in Table 1, the water-stability of the soils is quite uniform throughout the watershed the data are statistically identical   Slightly more variability is seen for both the mechanical stability (sonication) and the strength of the organic-matter-bound aggregates (peroxide)   The results also suggest that organic matter is more important for aggregation in the lower watershed soil, than in the upper regions   In the case of the upper watershed sample, the distribution of the < 2 μm mass is statistically indistinguishable whether sonication of peroxide is used as a dispersant   The lower watershed soils do show a significant difference between the two techniques   Similar mechanical stabilities, as determined by the sonication analyses, are seen for both the upper and lower soils

3 1 3 Figures
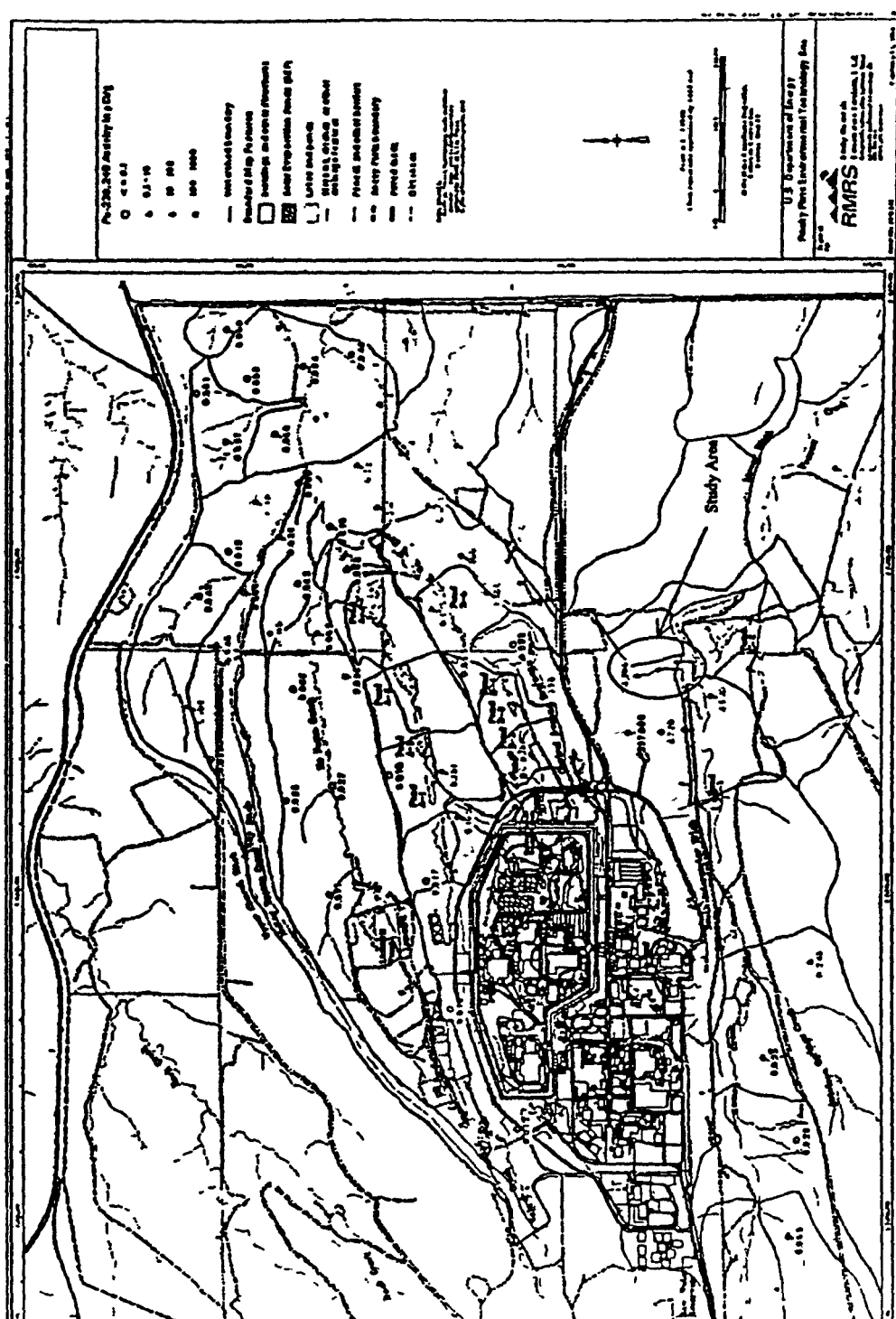
Figure 1 Watershed study site and 1998 surface soil sampling locations
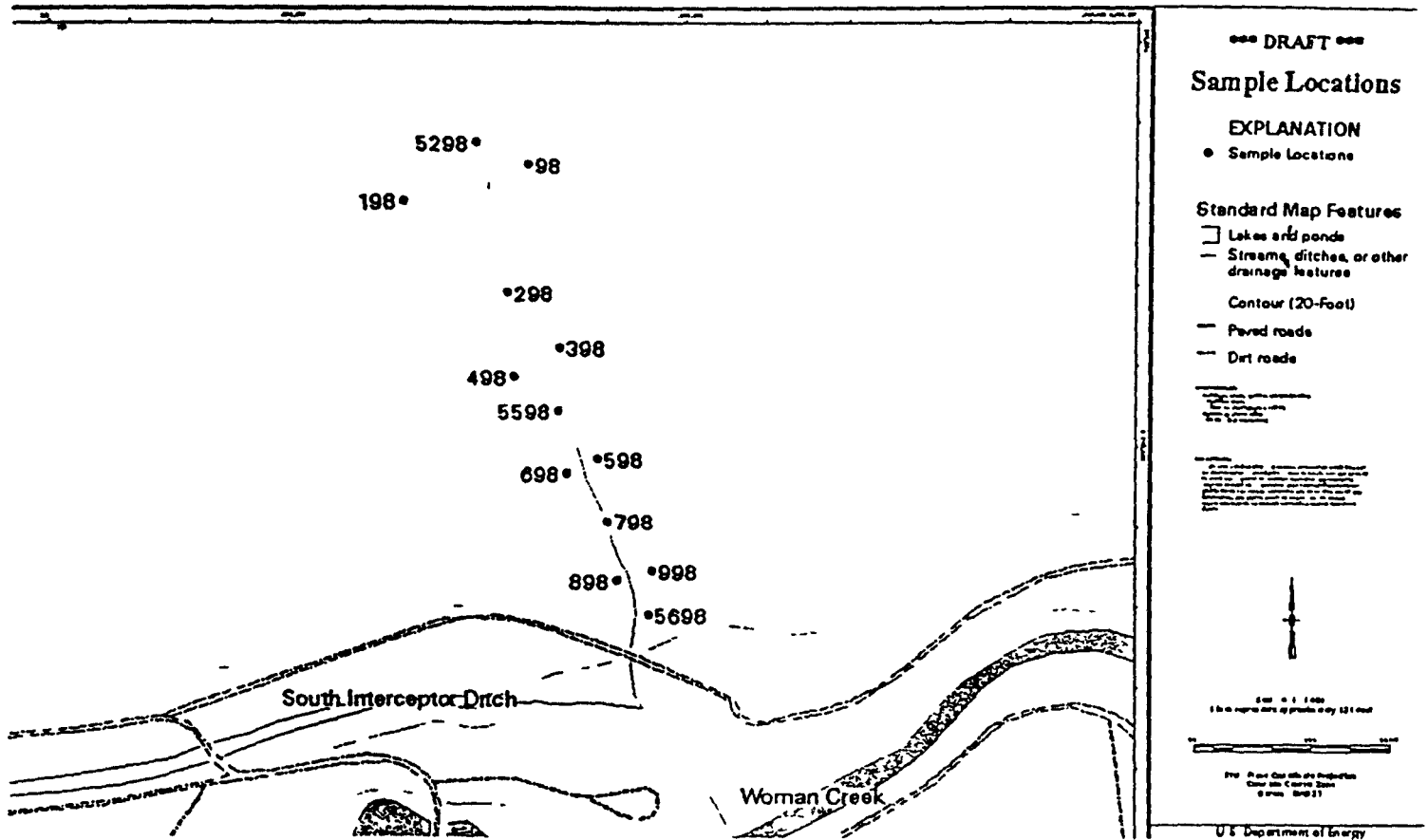
Figure 3   Soil sampling locations within the watershed study site
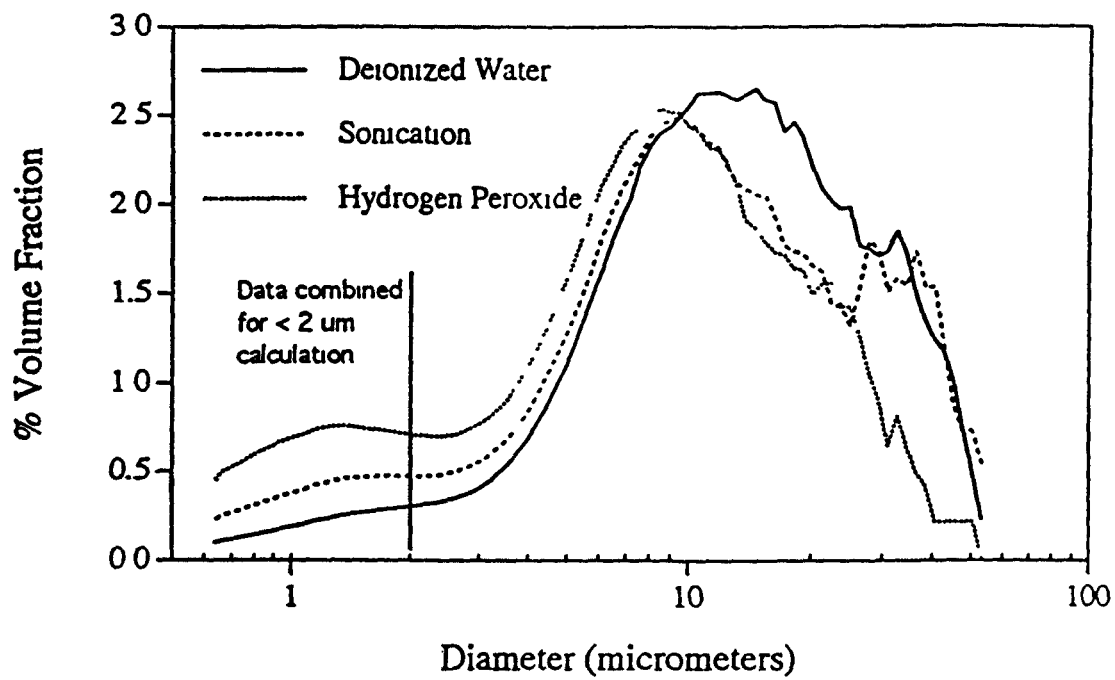
Figure 5 Volume distribution of the < 63 micrometer fraction of soil sample 898, obtained by single particle optical sizing (SPOS)

3 2 Soil Dispersion

3 2 1 Discussion

The affects of the different dispersants on the size distribution of various soil particle characteristics were examined The characteristics studied were 1) the mass of soil in each size fraction, 2) plutonium specific activity (pCi/g), 3) organic carbon content (%), 4) specific surface area (m2/g) and 5) iron content (ppm) The dispersants used for the upper watershed soil were deionized water (DI), ultrasonication(US), hexametaphosphate (HMP), hydrogen peroxide (H₂O₂), and citrate-dithionite-bicarbonate (CDB) For the lower soil, only DI, HMP, and US were used All the data on Pu 239/240 activity are presented in Table A-2

In the descriptions that follow, data is presented in two ways First the parameter value, on a per gram basis, of the various soil characteristics is presented Second, the percent distribution (inventory) of each characteristic is presented The inventories were determined by combining the distributions of soil mass with the parameter data. The entire data set is presented in Tables 2 through 9 In the accompanying figures, results from the deionized water dispersion will be used as a basis of comparison for the other dispersants The results of the iron analyses were not illustrative and are not presented in this report

Mechanical Stability A measure of the mechanical stability of the soil aggregates was obtained by examining the effect of ultrasonication on the size distributions Ultrasonication disrupts the soil aggregates without breaking the primary particles or chemically altering the soil The distribution of soil mass after sonication is compared to the water-stable aggregates in Figure 7 Most of the mass of the water-stable aggregates is in the sand fractions (> 53 micrometers) Sonication reduces the mass of particles in the sand fraction and increases the mass in the 10 to 0 45 μm fractions The effect is more pronounced for the upper watershed soil This result suggests that the lower watershed soils are more mechanically stable than the upper watershed soils

The affects of sonication on the plutonium activity (pCi ²³⁹ ²⁴⁰Pu/g) and plutonium inventory are presented in Figures 8 and 9, respectively The size distribution of plutonium activity is similar in the two watershed soils Sonication reduces the concentration of Pu in the larger size

a rupture of the tubing in the TFF, some of the solution in the 2 to 0 45 μm fraction was lost for one of the HMP dispersed samples Because the larger particles in this fraction had settled during the filtration, this resulted in a preferential loss of the finer particles in this fraction The mass in this fraction was corrected for this loss by mathematically adding mass to this fraction in order to make the measured percent recovery (84 9%) the same as the average for all samples (92 7%)

The plutonium activities and inventory for the HMP dispersed soils are shown in Figures 15 and 16. HMP dispersion causes both a greater decrease in the large fractions and an increase in the fine fractions as compared to sonication Most of the plutonium in the HMP-dispersed soil is in the 2 to 0 45 μm fraction, as is the case with particle mass More dissolved-phase, plutonium (< 10K Dalton) is observed for HMP dispersion as compared to sonication

HMP dispersion also results in a greater loss of surface area in the large particle sizes as compared to sonication (Figures 17 and 18) A quite dramatic increase in percent of the total surface area with decreasing particle size is observed This distribution is consistent with most of the BET-surface area being external surface area, suggesting a greater disruption of aggregates by HMP. The surface area for the 2 to 0 45 μm fraction in which some of the sample was lost may have been affected by the fact that the loss was preferentially of the smaller particles Therefore, instead of using the average obtained for this size fraction, only the unaffected sample result was plotted in Figures 17 and 18

Organic carbon concentrations and percent inventory are shown in Figures 19 and 20 Results for the organic carbon concentration are somewhat similar to the sonication results However, a greater reduction is seen for the 2000 to 200 and 25 to 10 μm fractions The percent of the total carbon present in the 2 to 0 45 μm fraction is higher for HMP dispersion The amount of organic carbon present in the

fraction The organic carbon inventory shows much more organic carbon in the less than 10 μm fractions, including dissolved organic carbon, than was present in the water-stable aggregates

Dissolution of iron oxides. The dissolution of iron oxide cements was attempted by the addition of citrate-dithionite-bicarbonate (CDB) to the soils The resulting distribution of soil mass, shown in Figure 21, is nearly identical to the HMP dispersion

The plutonium activity and total inventory distributions are shown in Figures 22 and 23 Results for the CDB treatment are again similar to HMP However considerably more Pu is seen in the dissolved (< 10K Dalton) and 53 to 25 μm fractions than was the case for HMP It was observed that, during the size fractionation procedure, the < 2 micrometer fraction, which was, kept in a closed container, became considerably reducing as indicated by both a sulfidic odor and the presence of a very black colloidal precipitate in the finest fractions We believe this was a result of sulfur-reducing bacteria present in the soil This result suggests that a significant amount of Pu can be released to solution (e g , in a colloid from) under extremely reducing conditions

Surface area results (Figures 24 and 25) show an increase in specific surface area with decreasing particle size Again, this is again consistent with a non-aggregated soil Most of the soil surface surface area is contributed by the 2 to 0 45 μm fraction

Because of the presence of citrate in the solutions, organic carbon was not determined

Comparison of the Percent Distributions of Various Soil Properties. Figures 28 –32 co-display the soil characteristics as a function of particle size fraction

Figure 28 shows the results for the water-stable aggregates Little Pu is released to the dissolved (< 10 K Dalton) phase under the conditions of the experiment The plutonium activity distribution most closely matches the mass and organic carbon distributions It does not appear that Pu activity is controlled by external surface area (i e , it does not follow a surface area distribution activity ∝ to particle diameter) Not enough mass was obtained for the < 0 45 μm fractions to allow surface area measurements However, we can assume that the specific (external) surface area will continue to increase with decreasing particle size Plutonium concentrations do not follow the increase in surface area in the fine fractions, nor does it correlate

3 2 2 Tables

Table 2: Soil Mass Distribution Data

Size Fraction (micron)	DI Water	HMP #1	HMP #1a*	HMP #2	Sonication	H2O2	CDB
Upper Watershed Soil (Mass in grams)							
2000 to 200	49 928	14.469	14.469	14 070	38 596	13.328	13 490
200 to 53	24.162	12 246	12.246	11.695	18 419	13 809	13 661
53 to 25	6.243	4.361	4.361	4.169	5 830	4.511	4 724
25 to 10	5 828	5 739	5.739	5 666	8 198	5 754	5 185
10 to 2	4.369	7.390	7.390	7.970	10 622	10.904	7 298
.2 to 0.45	2.537	*7.374*	*12.127*	12 661	9 904	8.043	11.742
0 45-10k	0.074	0.401	0.400	0.271	0 049	0.323	0 367
<10K	ND	ND	ND	ND	ND	ND	ND
total mass recovered	93 100	51.981	56.732	56 500	91.600	56.700	56 500
Mass Dispersed	100 756	61.200	61.200	60 348	100 380	61.811	60 596
% recovery	92 401	84 936	92.700	93 624	91 254	91.731	93 240

* Some of the fine fraction of HMP #1 2-0 45 micrometer was los

HMP #1 2-0 45 um was corrected by adjusting the amount in this fraction to obtain the average percent recovery (92 7%)

	DI Water 1	DI Water 2		HMP	Sonication
Lower Watershed Soil (Mass in grams)					
2000 to 200	42.145	20.708		12.635	43 953
200 to 53	28.074	12 615		10.762	19 126
53 to 25	6.102	2.746		3.015	4 348
25 to 10	6 862	3.953		4.367	7 546
10 to 2	5.378	4.400		5.881	8.846
2 to 0 45	4.150	2.139		10 284	5 266
0 45-10k	0.007	0 088		0.168	0 012
<10K	ND	ND		ND	ND
total mass recovered	92.719	46.650		47.200	89.000
Mass Dispersed	85.875	42.932		44.840	80 011
% recovery	92.619	92.031		95 000	89 900

ND· Not determined

Table 3 Soil Percent Mass Distribution Data

Size Fraction	DI Water	HMP #1	HMP #1a*	HMP #2	HMP Ave	HMP SD	Sonication	H2O2	CDB
Upper Watershed Soil									
2000 to 200	53 6	27 82	25 50	24 9	25 2	0 4	42 1	23 5	23 9
200 to 53	25 9	23 55	21.59	20 7	21 1	0 6	20 1	24 4	24 2
53 to 25	6.7	8.39	7,69	7 4	7 5	0 2	6 4	8 0	8 4
25 to 10	6.3	11 04	10 12	10 0	10 1	0 1	8 9	10 2	9 2
10 to 2	4 7	14 21	13 03	14 1	13 6	0 8	11 6	19 2	12 9
2 to 0 45	2.7	*14.18*	*21.38*	22.4	21.9	0 7	10 8	14 2	20 8
0 45-10k	0 1	0 77	0 71	0 5	0 6	0 2	0 1	0 6	0 7
<10K	ND	ND	ND	ND	ND		ND	ND	ND
% recovery	92 4	84 9	92 7	93 6			91 3	91 7	93 2

Some HMP#1   2-0 45 um was lost

* HMP 2-0 45 um was corrected by adjusting the amount in this fraction to obtain the average percent recovery (92 7%)

	DI Water #1	DI Water #2	DI Water Ave	DI SD	HMP	Sonication
Lower Watershed Soil						
2000 to 200	45 5	44 4	44 9	0 8	25 5	49 3
200 to 53	30 3	27 0	28 7	2 3	21 7	21 5
53 to 25	6.6	5 9	6 2	0 5	6 1	4 9
25 to 10	7 4	8 5	7 9	0 8	8 8	8 5
10 to 2	5 8	9 4	7 6	2 6	11 9	9 9
2 to 0 45	4 5	4 6	4 5	0 1	20 7	5 9
0 45-10k	0 0	0 2	0 1	0 1	0 4	0 0
<10K	ND	ND			ND	ND
% recovery	92 6	92 0			95 0	89 9

ND· Not determined

Table 6 Specific Surface Area (m2/g)

Upper Watershed Soil

Size Fraction (microns)	DI Water	HMP #1	HMP #2	HMP Ave.	HMP SD	Sonication	H2O2	CDB
2000 to 200	7.5	0.51	0.45	0.48	0 04	5 36	0.60	0 36
200 to 53	8.8	0.75	0.51	0 63	0 17	1 66	1.09	0 84
53 to 25	11.8	1.06	1.03	1 05	0 02	1.43	1.21	2 24
25 to 10	17.5	2.84	3.47	3 16	0 45	7.95	12 65	4 18
10 to 2	33.1	8.69	12.36	10.53	2 60	21 56	31 94	12 02
2 to 0 45	88 1	*42.29*	73 61	*73 61*	ND	84 33	69 06	30 45
Calculated Average SA	12.16	*7.96*	18.88	*18.20* ND		15.01	17.73	8.74
Unfractionated Soil SA	10.66							

Lower Watershed Soil

Size Fraction	DI Water #1	DI Water #2	DI Ave	SD	HMP	Sonication
2000 to 200	5 07	4.93	5.00	0 10	0 85	4.27
200 to 53	5.22	5.42	5.32	0 14	0.99	2.18
53 to 25	7.1	7.33	7.22	0.16	1.20	1 68
25 to 10	11.54	10.98	11.26	0 40	3.21	6 86
10 to 2	20.85	23.44	22.15	1 83	12 51	19.90
2 to 0 45	54.17	59 31	56 74	3 63	43.24	51.04
Calculated Average SA	8 84	9.95	9.39	0 78	11 81	8 24
Unfractionated Soil SA	6.88					

ND Not determined

Table 7. Inventory of the percent of the total specific surface area in each size fraction.

Upper Watershed Soil

Size Fraction (microns)	DI Water	HMP ave	HMP SD	Sonication	H2O2	CDB
2000 to 200	33 21	0 90	0 43	15 04	0 80	0 98
200 to 53	18.78	1 03	0 66	2 22	1 50	2 32
53 to 25	6 49	0.58	0 25	0 61	0 54	2 14
25 to 10	9.00	2 25	0 57	4 74	7 24	4 39
10 to 2	12.76	9 84	0 86	16 65	34 65	17 76
2 to 0 45	19.76	85.41	2 77	60 73	55 27	72 40

Lower Watershed Soil

Size Fraction	DI Water ave.	DI Water SD		HMP	Sonication
2000 to 200	24 03	2.87		1 93	25 60
200 to 53	16 31	2 22		1 91	5 69
53 to 25	4 81	0 67		0 65	1 00
25 to 10	9 51	0 22		2 52	7 06
10 to 2	17 95	6 04		13 20	24 01
2 to 0 45	27 39	0 06		79 79	36 65
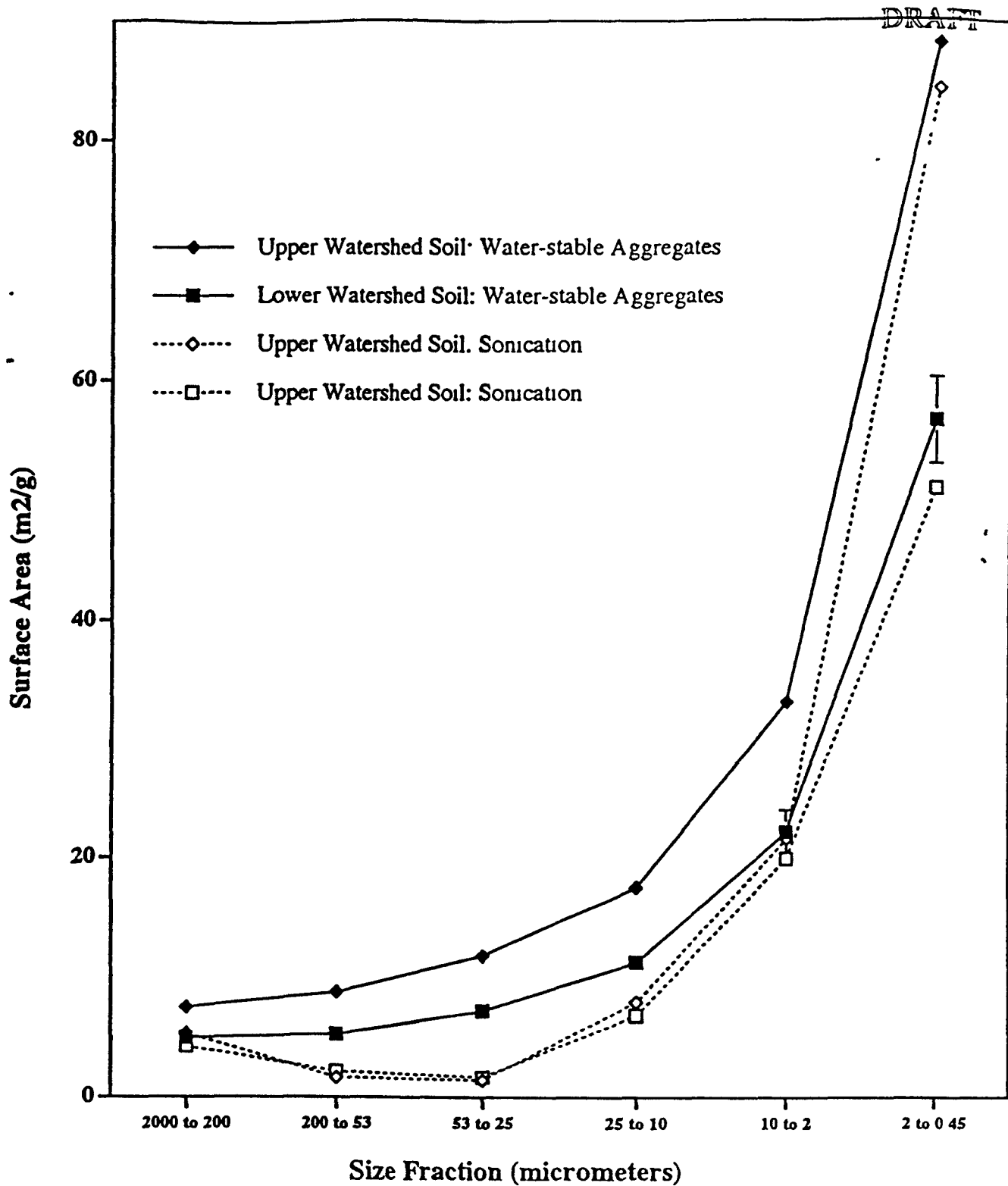
Figure 10 Soil aggregate mechanical stability Influence of sonication on the specific surface area (m^2/g) of the upper and lower watershed soils
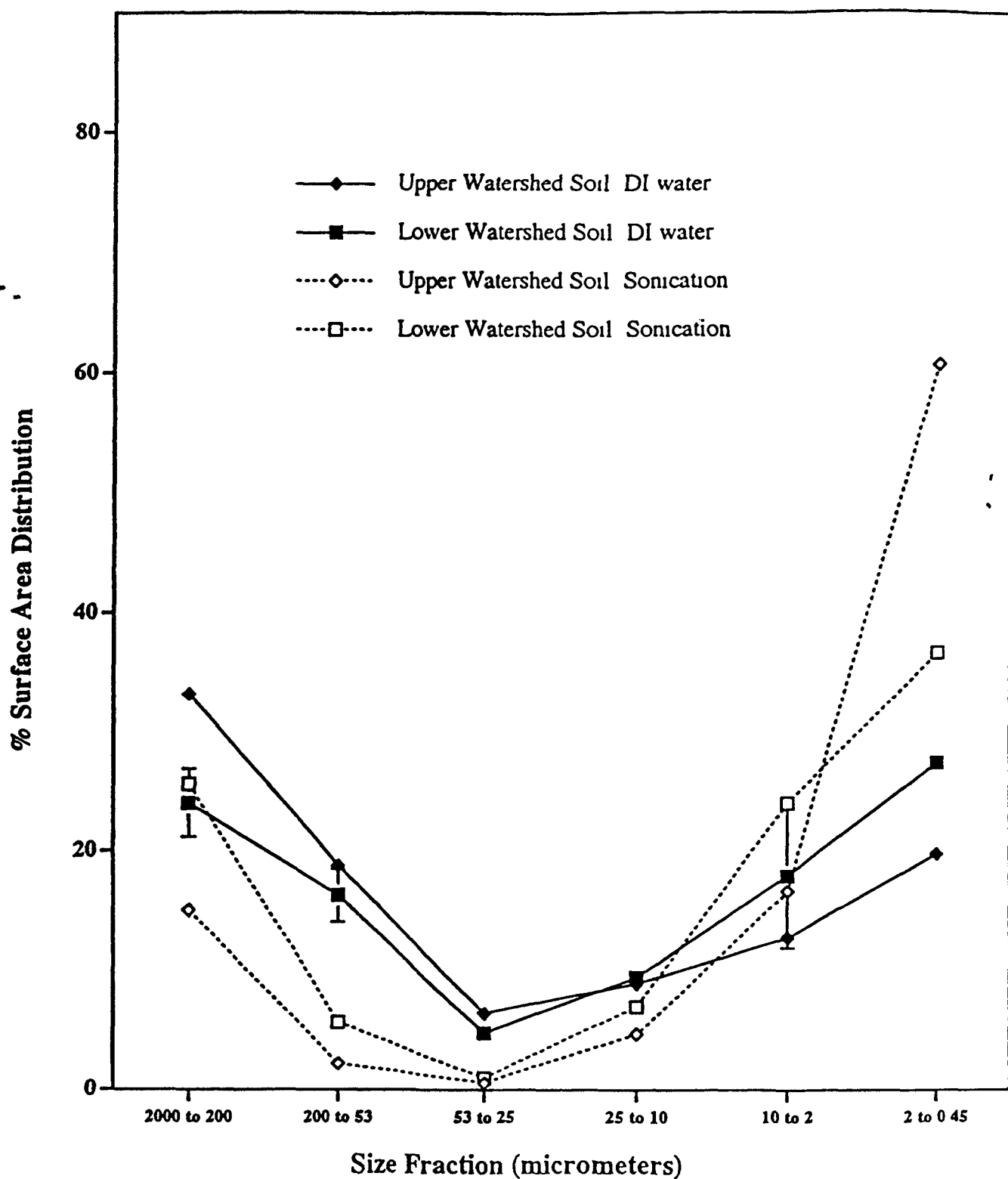
Figure 11. Soil aggregate mechanical stability Influence of sonication on the percent specific surface area inventory of the upper and lower watershed soils
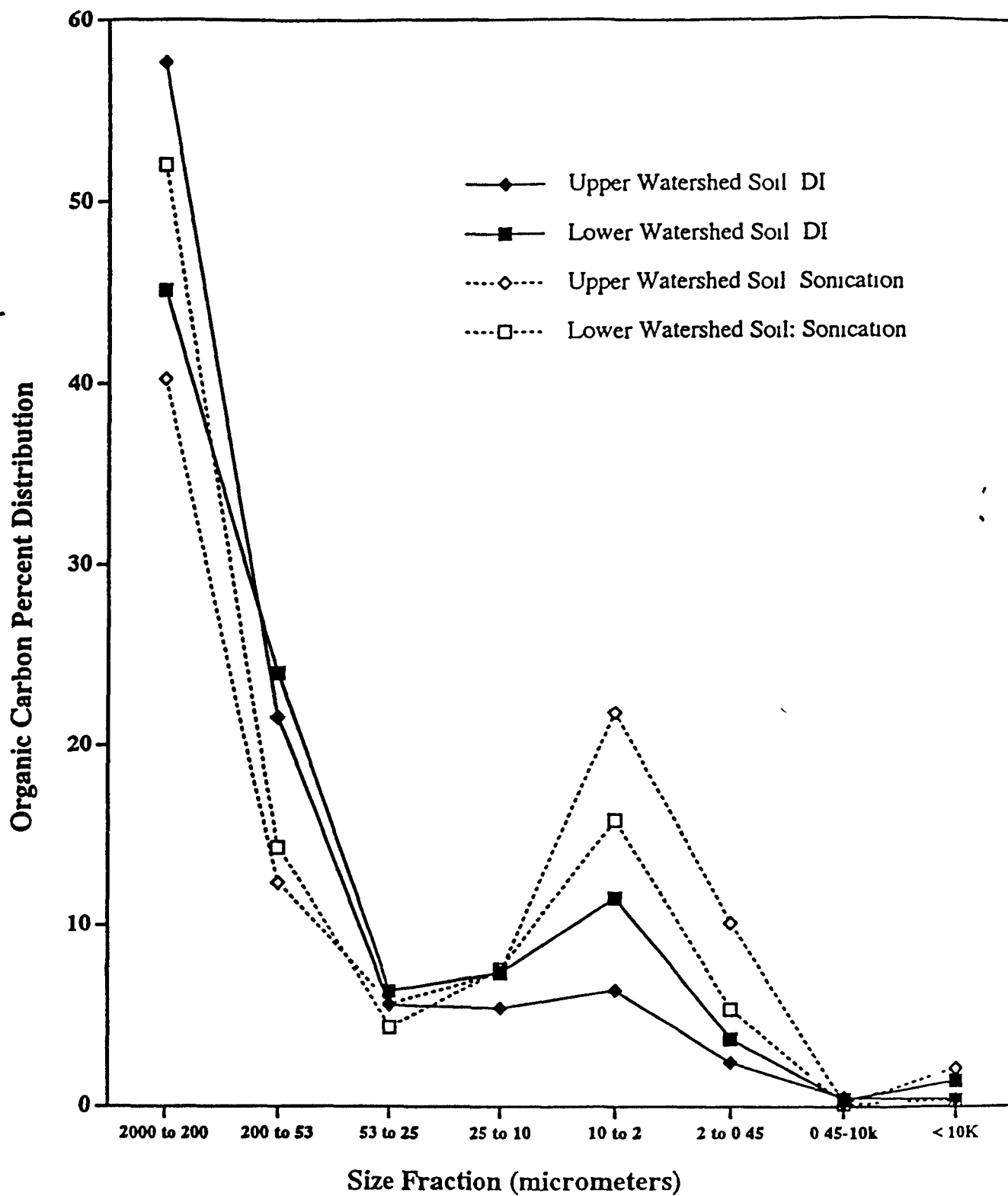
Figure 13   Soil aggregate mechanical stability   Influence of sonication on the percent organic carbon inventory of the upper and lower watershed soils
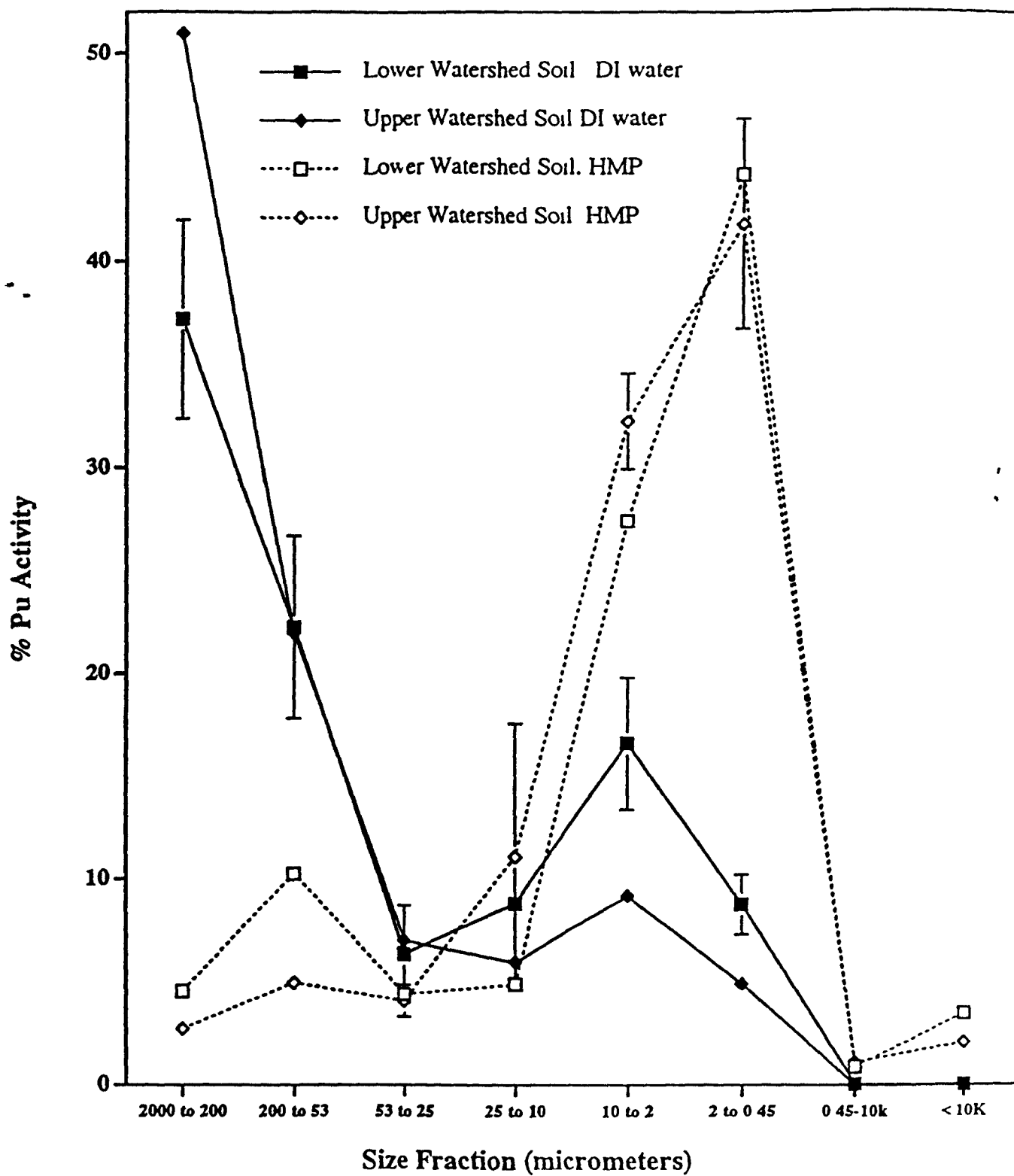
Figure 16 Soil aggregate chemical stability Influence of hexametaphosphate on the the Pu 239/240 percent activity inventory of the upper and lower watershed soils
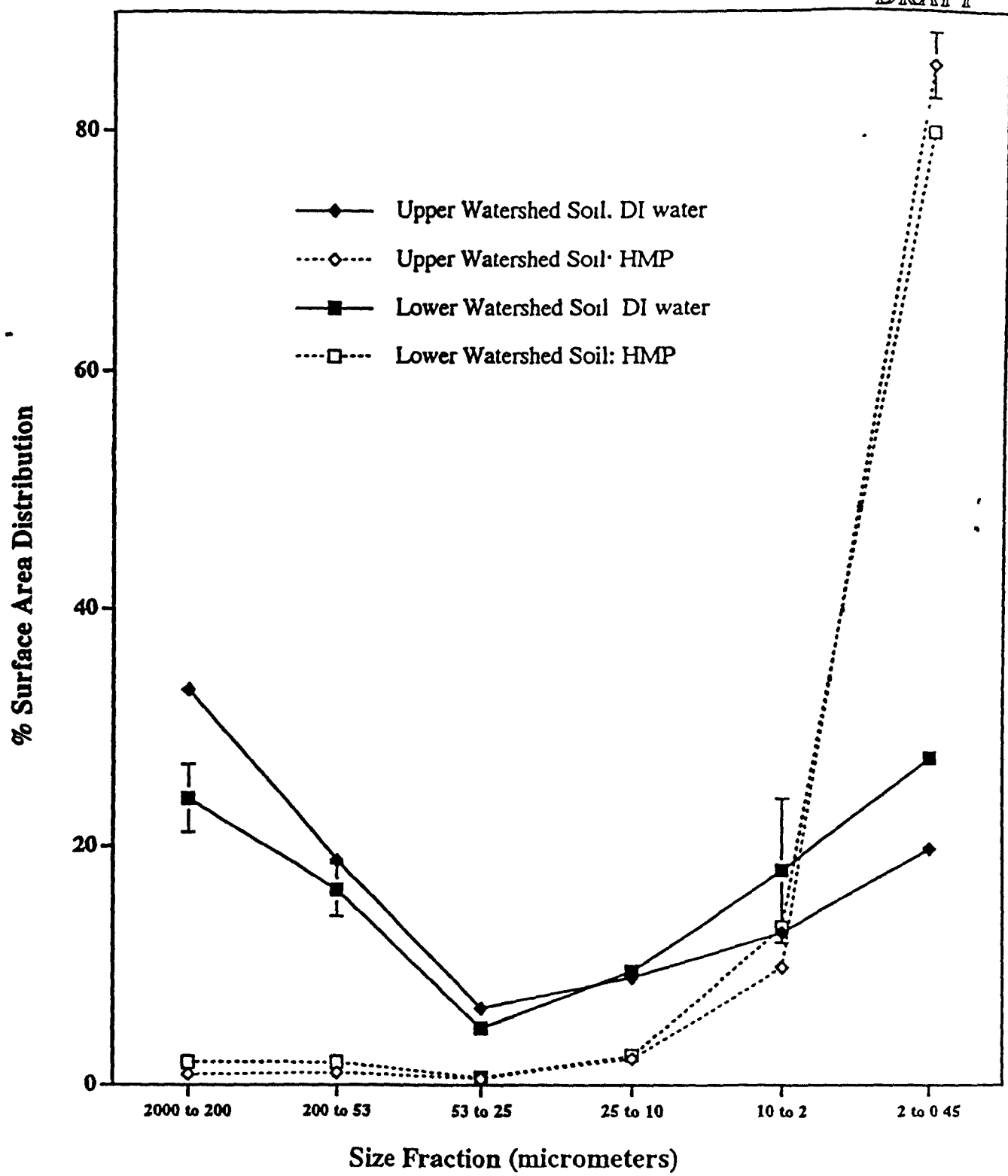
Figure 18 Soil aggregate chemical stability Influence of hexametaphosphate on the percent specific surface area inventory of the upper and lower watershed soils
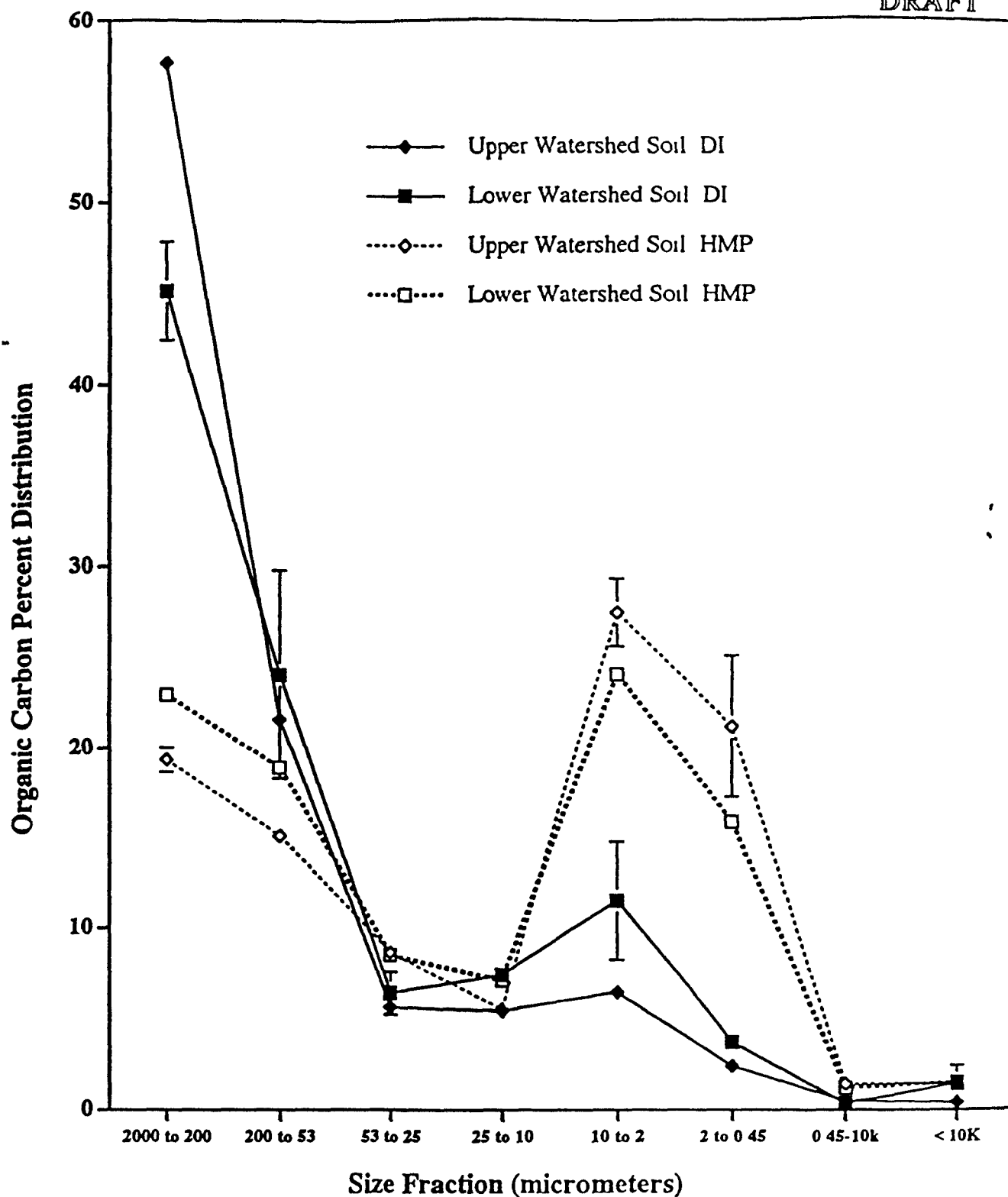
Figure 20 Soil aggregate chemical stability Influence of hexametaphosphate on the percent organic carbon inventory of the upper and lower watershed soils
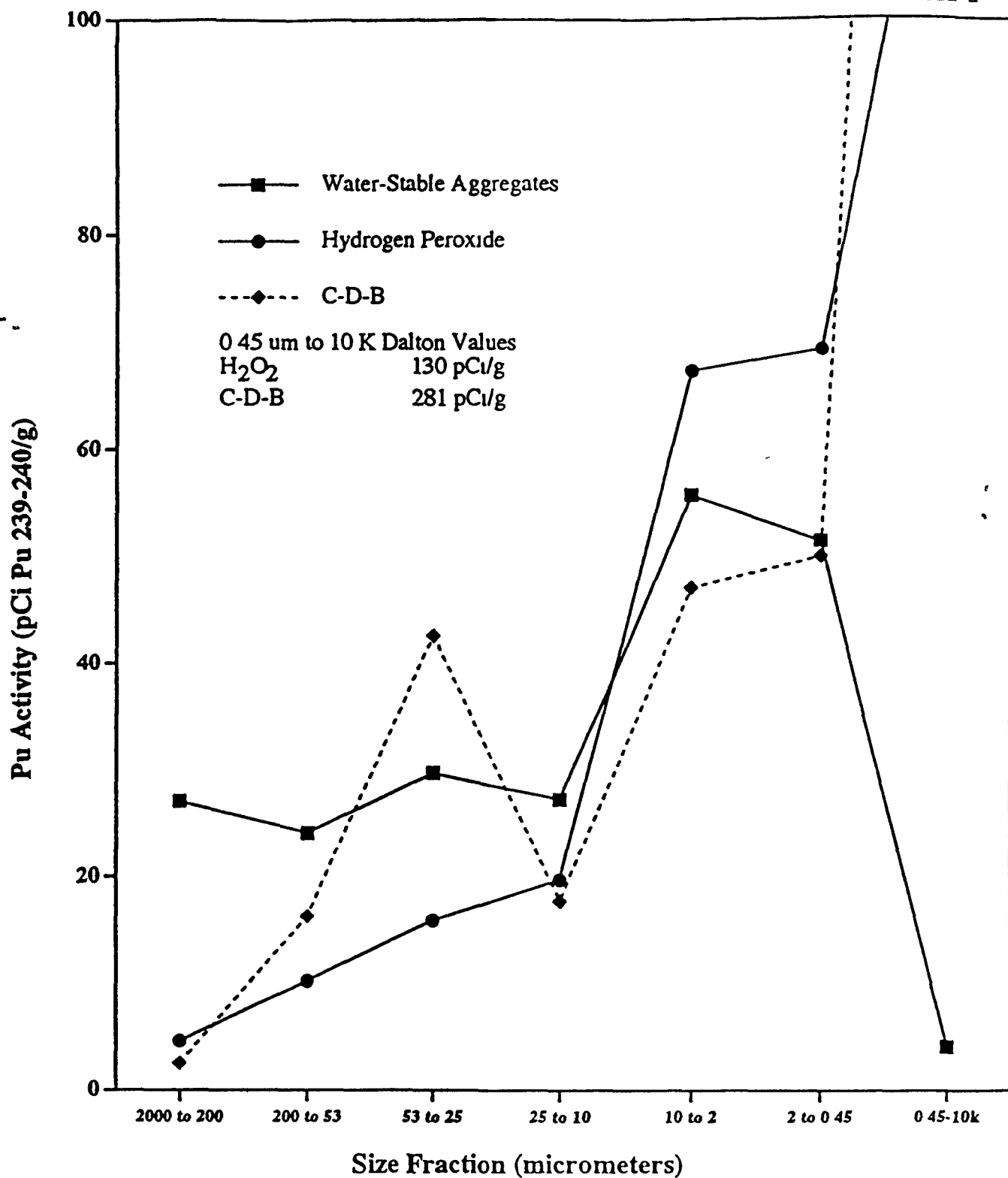
Figure 22 Soil aggregate stability due to organic matter and iron oxides Influence of hydrogen peroxide and citrate-dithionite-bicarbonate on the Pu 239/240 activities (pCi Pu/g) of the upper watershed soil
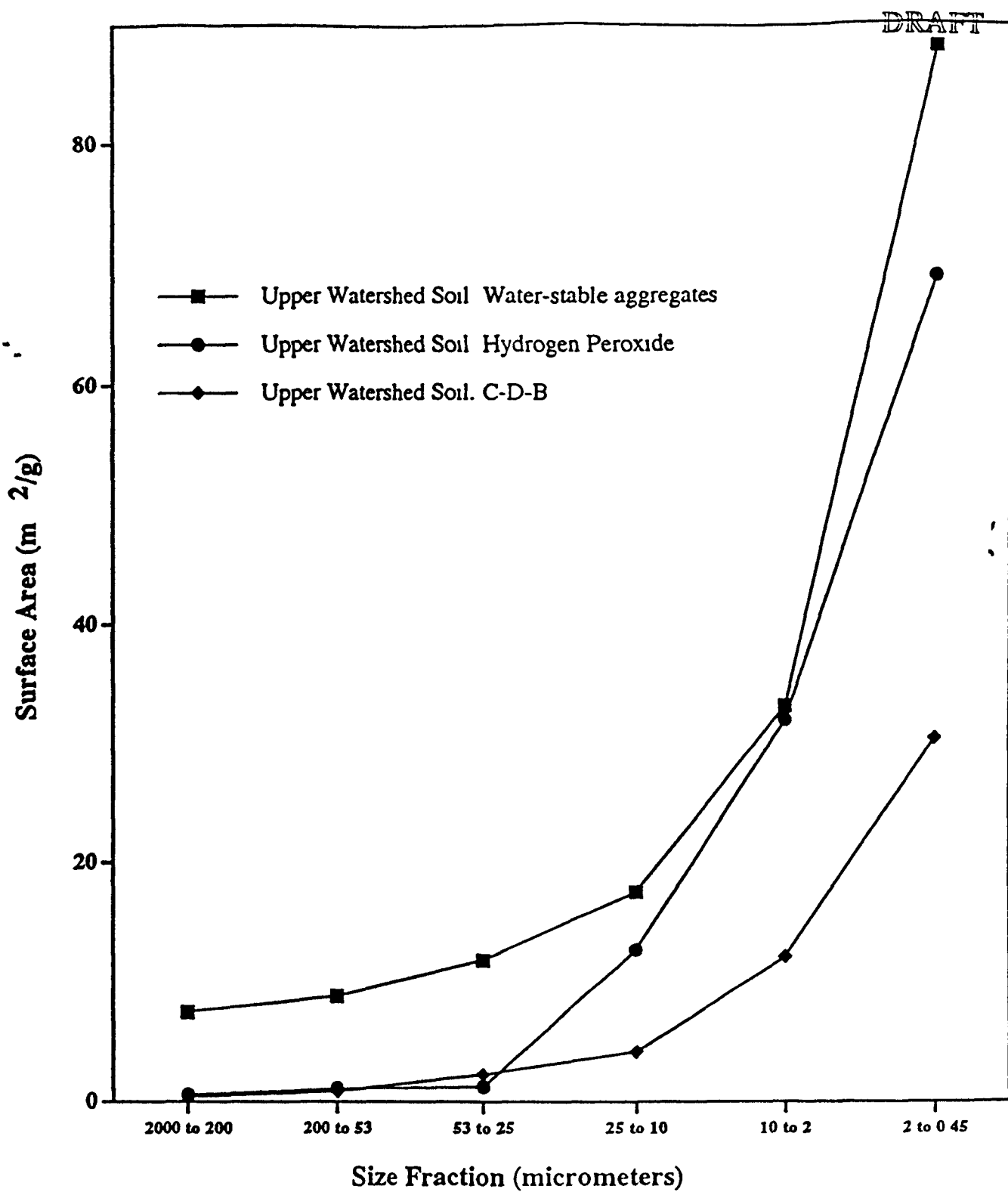
Figure 24   Soil aggregate stability due to organic matter and iron oxides Influence of hydrogen peroxide and citrate-dithionite-bicarbonate on the specific surface area of the upper watershed soil
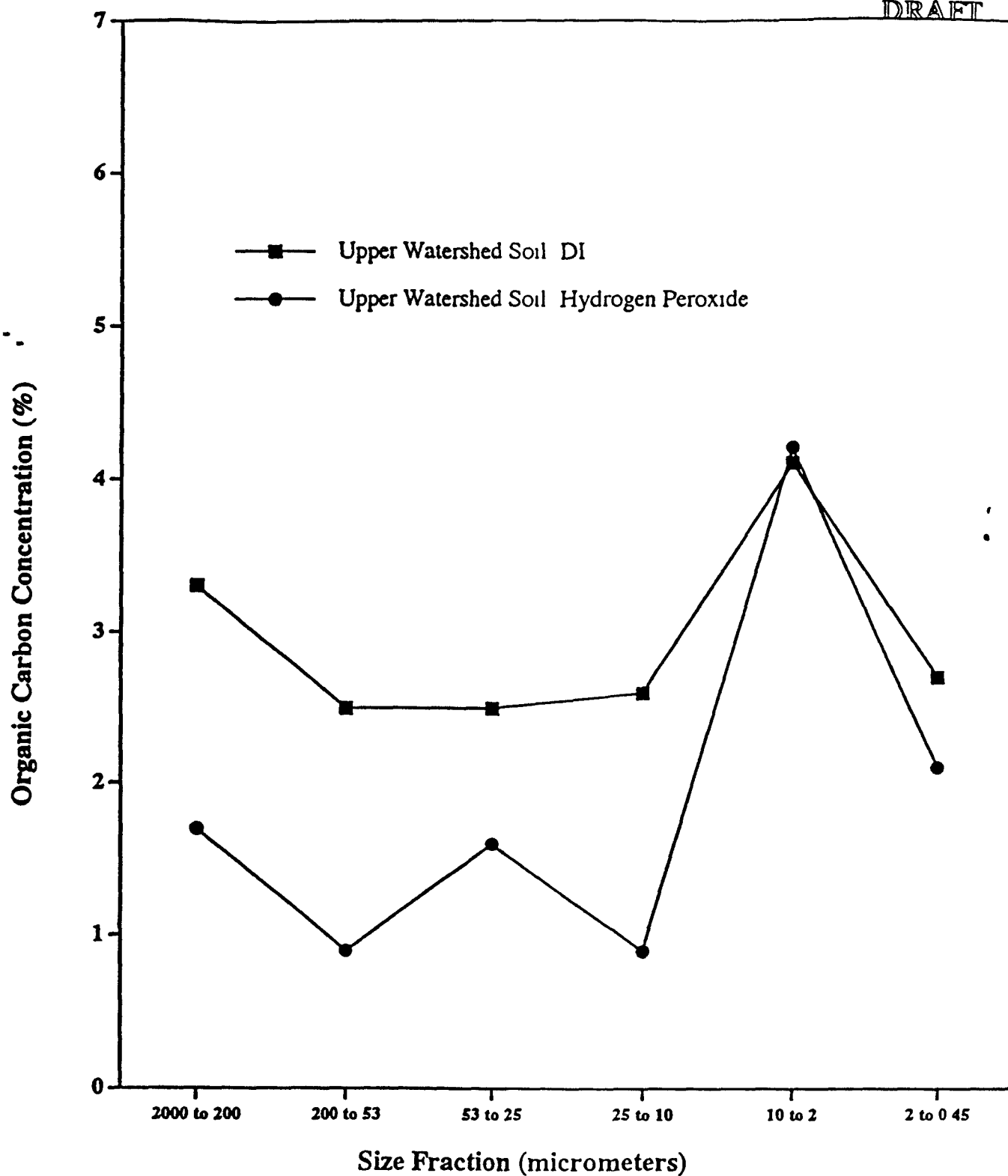
Figure 26   Soil aggregate stability due to organic matter Influence of hydrogen peroxide on the organic carbon concentration (%) of the upper watershed soil

25
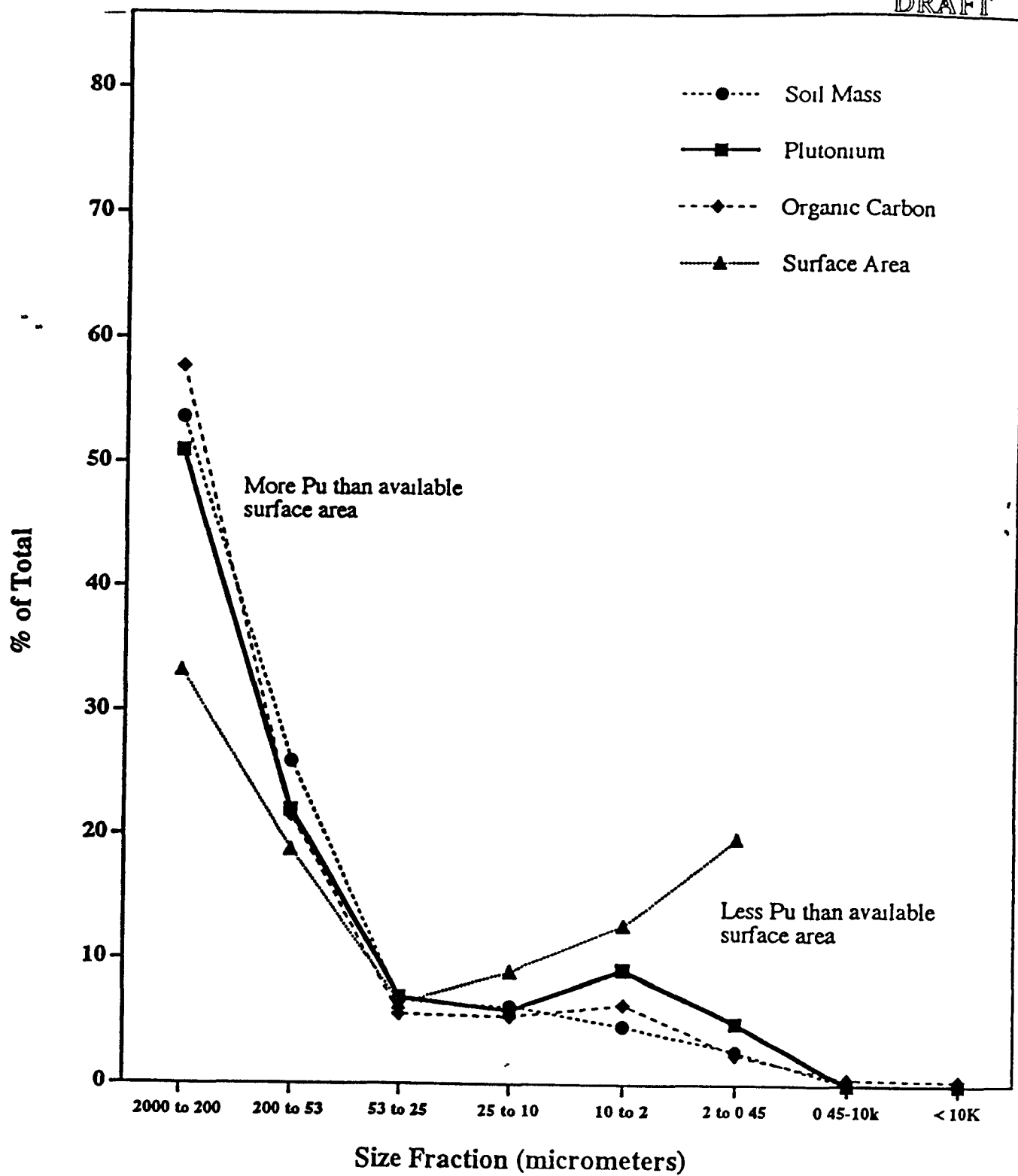
Figure 28   Comparison of the soil characteristics of the water-stable aggregatres in the upper watershed soil

as compared to the initial distribution in the runoff sample   In general, the 'dissolved' activity ranges from 0 4 to 0 8 pCi Pu/L, which is approximately the limit of Pu solubility, depending, of course, on the selection of the controlling Pu solid phase   A high value was obtained for the sonication sample, which is suspected to be in error   Also it must be noted that very low tracer yields were obtained for three of the samples   Sedimentation or aggregation will not remove dissolved plutonium   Adsorption processes will instead be required to effect the removal of plutonium transported in this fraction

The relative mobility of plutonium in surficial runoff is illustrated in Figure 36 where the percent distribution of plutonium in the runoff and the deionized water dispersion of the lower watershed soil is compared   Addition of water to the soil samples released a very small , percentage of plutonium to the less than 2 μm size fractions   In contrast, about 80 % of the plutonium in the runoff sample is present in the less than 2 μm fraction   The results suggest that plutonium in the < 2 0 μm fraction is far more mobile than plutonium in the larger fractions, at least under the conditions of the single runoff event that was sampled   Further work should be done to confirm these results, especially in light of the previously discussed uncertainties in the sample collection and size fractionation methods

Table 11 Pu 239 240 Data for the Runoff Sample

Pu 239/240 (pCi)	Deionized Water	Tracer % Yield	Sonication	Tracer % Yield	Hydrogen Peroxide	Tracer % Yield	Hexametaphosphate	Tracer % Yield	C-D-B	Tracer % Yield
>25 micron	0 256	35 6	0 035	40 0	0 091	34 9	0 057	33 7	0 013	26 3
25-10 micron	0 000	41 6	0 000	37 8	0 000	22 9	0 000	27 6	0 000	32 1
10-2 micron	0 205	38 4	0 328	45 1	0 000	29 3	0 000	26 3	0 148	18 3
2-0 45 micron	0 577	28 6	0 643	48 8	1 606	37 5	0 596	36 2	*1 292*	*4 4*
0 45-10K Dalton	0 240	38.2	0 276	38 6	0 233	31 9	0 173	21 9	0 215	29 2
<10K Dalton	*0 934*	*0 3*	*3 089*	*2 6*	*1 229*	*0 8*	1 685	33 1	1 466	23 0
Total Pu	2 212		4 370		3 159		2 510		3 134	
Sample Volume (Liters)	1 894		1 795		1 989		1 989		1 797	
Pu 239/240 pCi/L										
>25 micron	0 135		0 019		0 046		0 029		0 007	
25-10 micron	0 000		0 000		0 000		0 000		0 000	
10-2 micron	0 108		0 183		0 000		0 000		0 082	
2-0 45 micron	0 305		0 358		0 807		0 300		0 719	
0 45-10K Dalton	0 127		0 154		0 117		0 087		0 120	
<10K Dalton	0 493		1 721		0 618		0 847		0 816	
Total pCi Pu/L	1 168		2 435		1 588		1 262		1 745	

Average pCi Pu/L   Standard Deviation
1 640   0 503

* *Results in italics have very low tracer yields*

Average Blank pCi Pu/L   Standard Deviation
0 059   0.046

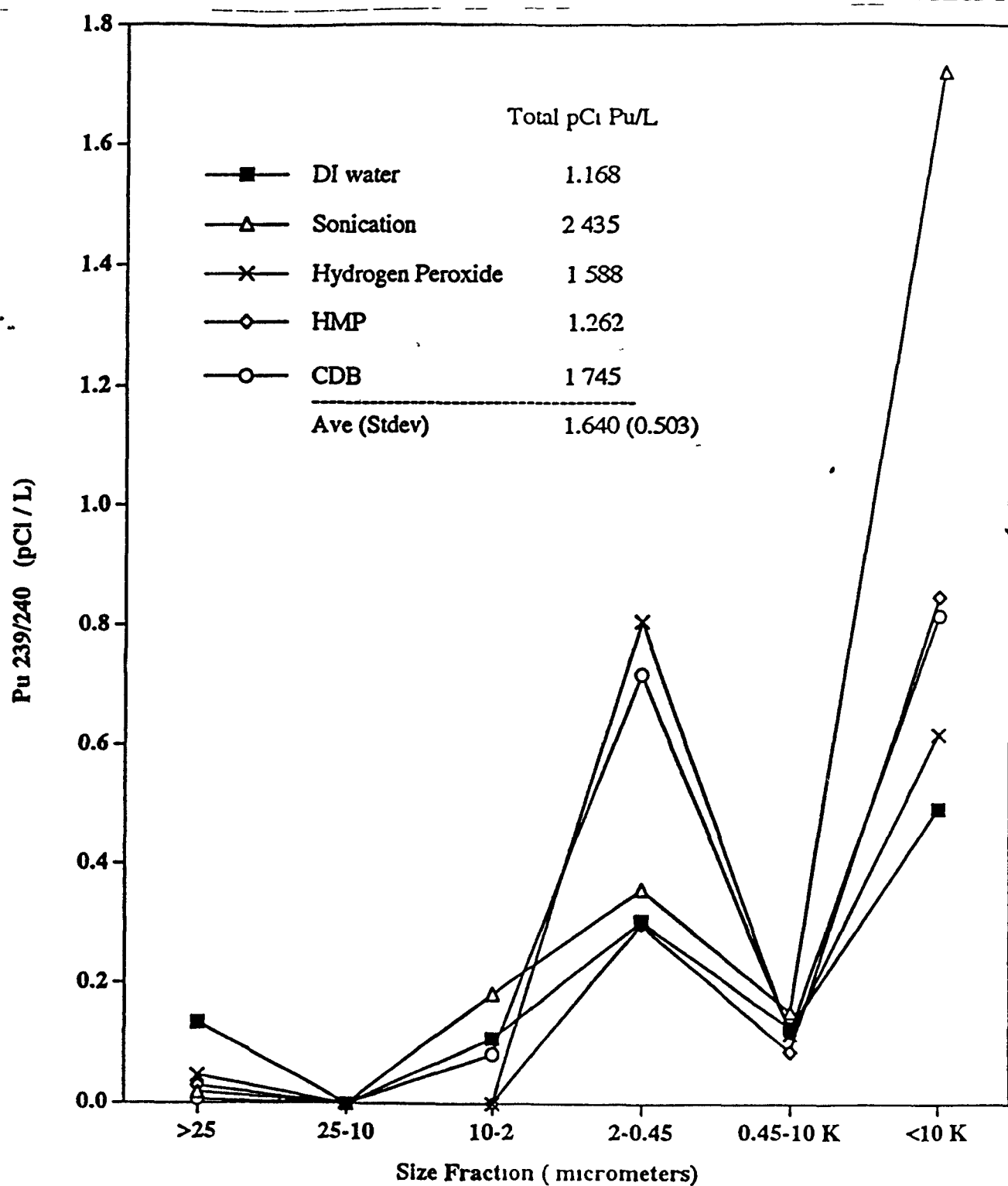| % Pu Distribution	Deionized Water	Sonication	Hydrogen Peroxide	Hexametaphosphate	C-D-B
>25 micron	11 6	0.8	2 9	2 3	0 4
25-10 micron	0 0	0 0	0 0	0 0	0 0
10-2 micron	9 3	7 5	0 0	0 0	4 7
2-0 45 micron	26 1	14 7	50 8	23 7	*41 2*
0 45-10K Dalton	10 8	6.3	7 4	6 9	6 9
<10K Dalton	*42 2*	*70 7*	*38 9*	67 1	46 8

Figure 34. Size distribution of Pu 239/240 activity (pCi Pu/L) in the runoff sample collected from GS-42 on April 30, 1999
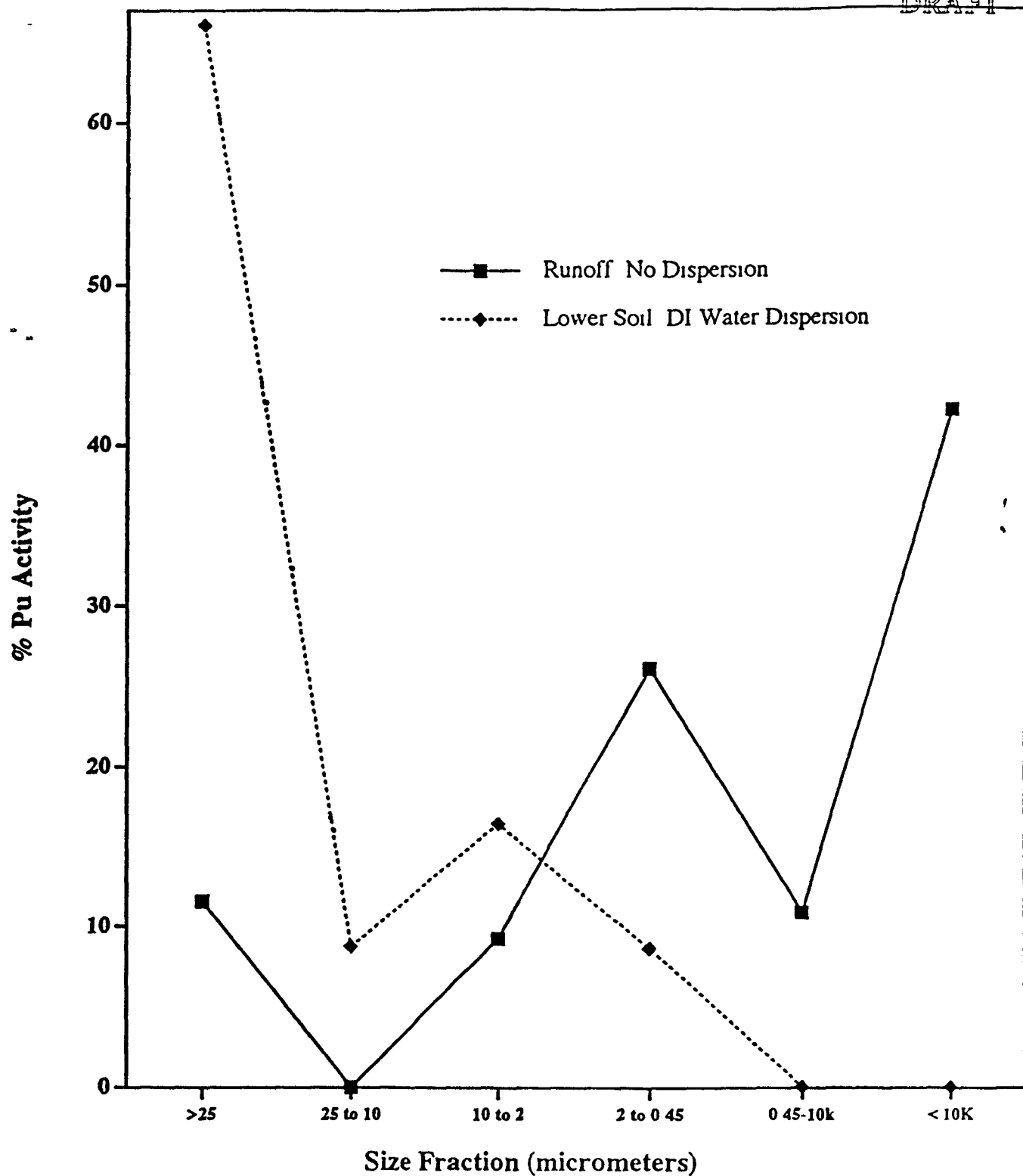
Figure 36 Comparison of the percent plutonium activity size distributions of the runoff sample versus the water-stable aggregates from the lower watershed soil

Table A-2 All Pu 239/240 and Percent Organic Carbon Data for Watershed Soils

SAMPLE DESCRIPTION	MASS OF SAMPLE (G)	TRACER Bq ADDED	TRACER Pu-239/240 COUNTS	COUNTS	COUNT TIME (m)	SAMPLE ACTIVITY (Bq/G)	COUNTING UNCERT (1s)(Bq/G)	SAMPLE ACTIVITY (pCi/G)	COUNTING UNCERT (1s)(pCi/G)	DETECTOR EFFICIENCY	CHEMICAL YIELD %	SAMPLE SPECIFIC MDA (pCi/g)	Volumetric Multiplier Pu data	Volume Corrected pCi Pu/g	Blank Subtracted pCi Pu/g
TOTAL SOIL 098	0.7141	0.2021	1179	3255	1000	0.78134793	0.02655889	21.1174906	0.71780711	0.3152	30.8467939	0.08503912	1.03413988	21.8384393	21.7569633
TOTAL SOIL 198	1.1664	0.2021	17571	10828	1000	1.06781321	0.02746388	28.8597876	0.74226629	0.3152	45.9693104	0.03493593	1.04091739	30.0406547	29.9591787
TOTAL SOIL 298	1.3487	0.2021	1505	6637	1000	0.66082471	0.01886677	17.860109	0.50991232	0.3152	39.3761025	0.03527279	1.02952961	18.3875115	18.3060355
TOTAL SOIL 398	1.3743	0.2021	2996	12425	1000	0.60987293	0.01241299	16.4830356	0.33548597	0.3152	78.3859157	0.01738875	1.03333035	17.0324209	16.9509449
TOTAL SOIL 498	1.2664	0.2021	1361	4508	1000	0.52839274	0.01634866	14.286276	0.44185531	0.3152	35.6085552	0.04153963	1.04587611	14.9416748	14.8601988
TOTAL SOIL 598	1.1398	0.2021	1698	4089	1000	0.42698939	0.01232726	11.5402423	0.33316876	0.3152	44.4256625	0.03699349	1.03745243	11.9724524	11.8909764
TOTAL SOIL 698	1.6694	0.2021	1787	5025	1000	0.34042184	0.00937615	9.20058114	0.25340914	0.3152	46.7542161	0.02399975	1.03331416	9.50709074	9.42561474
TOTAL SOIL 798	1.262	0.2021	1433	3032	1000	0.33883633	0.01086208	9.15772941	0.29356944	0.3152	37.4923289	0.03959005	1.03235041	9.45398572	9.37250972
TOTAL SOIL 898	1.6538	0.2021	2332	7525	1000	0.39433133	0.00934579	10.6575929	0.25258874	0.3152	61.0133363	0.01856436	1.03658926	11.0475463	10.9660703
TOTAL SOIL 998	1.4491	0.2021	2117	6237	1000	0.41088742	0.01033527	11.1050543	0.27933131	0.3152	55.3881788	0.02333847	1.0336924	11.4792102	11.3977342
TOTAL SOIL 5298	0.7984	0.2021	2167	20588	1000	2.40492221	0.05431289	64.9978326	1.4679145	0.3152	56.6963549	0.04138207	1.02718713	66.764937	66.683461
TOTAL SOIL 5698	1.2857	0.2021	1815	3437	1000	0.29766623	0.00863702	8.0450253	0.2334327	0.3152	47.4867947	0.03068141	1.03839199	8.35388982	8.27241382
PROCEDURE BLANK	1	0.2021	1676	25	1000	0.00301462	0.0006074	0.08147608	0.0164163	0.3152	43.850065	0.04271866	1	0.081476	0.081476

	% OC	Std Dev	% IC
TOTAL SOIL 098	2.90	0.03	ND
TOTAL SOIL 198	3.10	0.30	ND
TOTAL SOIL 298	3.72	0.12	ND
TOTAL SOIL 398	3.03	0.12	ND
TOTAL SOIL 498	3.98	0.16	0.13
TOTAL SOIL 598	3.81	0.04	ND
TOTAL SOIL 698	3.68	0.17	ND
TOTAL SOIL 798	3.52	0.06	ND
TOTAL SOIL 898	3.71	0.07	ND
TOTAL SOIL 998	4.63	0.18	ND

ND - not detected

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Table A-4 All Pu 239/240 data for runoff sample

SAMPLE DESCRIPTION	TRACER Bq ADDED	TRACER COUNTS	Pu-239/240 COUNTS	COUNT TIME (m)	SAMPLE ACTIVITY (Bq)	COUNTING UNCERT (1σ)(Bq)	SAMPLE ACTIVITY (pCi)	COUNTING UNCERT (1σ)(pCi)	DETECTOR EFFICIENCY	CHEMICAL YIELD %	SAMPLE SPECIFIC MDA (pCi/g)	VOLUMETRIC MULTIPLIER	CORRECTED TOTAL PU ACTIVITY pCi Pu	BLANK SUBTRACTED PU ACTIVITY pCi Pu
RUNOFF DI > 25 um	0.2023	1360	78	1000	0.01160	0.00135	0.31358	0.00135	0.31520	35.54721	0.05270	1.02592744	0.321711115	0.256474399
RUNOFF SONIC > 25 um	0.2023	1529	27	1000	0.00357	0.00069	0.09655	0.01874	0.31520	39.86448	0.04687	1.0352263	0.099950376	0.034714059
RUNOFF CDB > 25um	0.2023	1005	14	1000	0.00282	0.00076	0.07617	0.02050	0.31520	26.26893	0.07131	1.0304474	0.078484071	0.013247533
RUNOFF HMP > 25 um	0.2023	1288	28	1000	0.00440	0.00084	0.11886	0.02271	0.31520	33.66530	0.05584	1.02795887	0.122183238	0.056946722
RUNOFF H2O2 > 25 um	0.2023	1338	37	1000	0.00560	0.00093	0.15142	0.02324	0.31520	34.91991	0.05364	1.0312641	0.156156077	0.09091956
PROCEDURE BLANK	0.2023	863	10	1000	0.00234	0.00075	0.06336	0.02015	0.31520	22.55680	0.06304	1.02969318	0.065236517	0
RUNOFF CDB 25 10 um	0.052	316	44	1000	0.00724	0.00117	0.19569	0.03149	0.31520	32.13263	0.05830	1.02731535	0.201034483	-0.036009601
RUNOFF CDB 10-2 um	0.052	180	48	1000	0.01387	0.00225	0.37477	0.06088	0.31520	18.30340	0.10234	1.02798151	0.385261152	0.148217068
RUNOFF CDB 2-0 45 um	0.052	43	45	1000	0.05442	0.01161	1.47077	0.31365	0.31520	4.37248	0.42841	1.03949784	1.528848923	1.291804039
RUNOFF CDB 0 45 um 10K De	0.052	287	90	1000	0.01631	0.00197	0.44072	0.05324	0.31520	29.18375	0.06419	1.02647645	0.452387701	0.215343817
RUNOFF H2O2 25-10 um	0.052	225	31	1000	0.00718	0.00137	0.19363	0.03710	0.31520	22.87925	0.08187	1.03310581	0.200043832	-0.037000232
RUNOFF H2O2 10-2 um	0.052	288	29	1000	0.00524	0.00102	0.14152	0.02737	0.31520	29.28544	0.06396	1.02753782	0.145413428	-0.091630858
RUNOFF H2O2 2-0 45 um	0.052	369	472	1000	0.06651	0.00462	1.79770	0.12492	0.31520	37.52196	0.04992	1.02512894	1.842872587	1.605828503
RUNOFF H2O2 0 45 um-10K De	0.052	314	102	1000	0.01689	0.00193	0.45653	0.05203	0.31520	31.92926	0.05867	1.02965172	0.470069481	0.233025399
PROCEDURE BLANK	0.052	540	58	1000	0.00856	0.00124	0.23148	0.03338	0.31520	34.57308	0.05418	1.02404449	0.237044084	0
RUNOFF SONIC <10K De	0.0524	28	58	1000	0.11288	0.02678	3.05031	0.72389	0.31520	2.82364	0.71198	1.02397799	3.12954969	3.085598153
RUNOFF DI <10K De	0.0524	8	7	1000	0.04585	0.02373	1.23919	0.64134	0.31520	0.80727	2.32042	1.02485868	1.26899253	1.229040993
RUNOFF DI <10K De	0.0524	3	2	1000	0.03493	0.03189	0.94414	0.86188	0.31520	0.30273	6.18779	1.03282847	0.975137978	0.934186441
RUNOFF HMP <10K De	0.0524	328	391	1000	0.06248	0.00468	1.68823	0.12641	0.31520	33.09825	0.05660	1.02209689	1.725536332	1.684584795
RUNOFF CDB <10K De	0.0524	228	283	1000	0.05355	0.00499	1.44727	0.13482	0.31520	23.00792	0.08142	1.04111757	1.506780454	1.465828918
PROCEDURE BLANK	0.0524	318	9	1000	0.00148	0.00050	0.04008	0.01355	0.31520	32.08916	0.05838	1.02170539	0.040951537	0
RUNOFF DI 25 10 um	0.0524	412	20	1000	0.00254	0.00058	0.06875	0.01574	0.31520	41.57464	0.04506	1.02990066	0.070803911	-0.031457237
RUNOFF DI 10-2 um	0.0524	381	80	1000	0.01100	0.00135	0.29737	0.03657	0.31520	38.44645	0.04872	1.03354266	0.30734245	0.205081302
RUNOFF DI 2-0 45 um	0.0524	263	132	1000	0.02444	0.00258	0.66057	0.06962	0.31520	26.55734	0.06559	1.02786851	0.678975625	0.576714478
RUNOFF DI 0 45 um 10K De	0.0524	379	89	1000	0.01231	0.00145	0.33257	0.03917	0.31520	38.24463	0.04898	1.02832609	0.341987931	0.239726784
RUNOFF SONIC 25 10 um	0.0524	375	10	1000	0.00140	0.00045	0.03777	0.01210	0.31520	37.84099	0.04950	1.02955991	0.03888208	-0.063379068
RUNOFF SONIC 10-2 um	0.0524	447	131	1000	0.01538	0.00153	0.41504	0.04124	0.31520	45.10646	0.04153	1.03611179	0.430033292	0.327772144
RUNOFF SONIC 2-0 45 um	0.0524	464	245	1000	0.02652	0.00208	0.71689	0.05621	0.31520	46.84011	0.03835	1.03695838	0.744814296	0.642553148
RUNOFF SONIC 0 45 um-10K De	0.0524	383	100	1000	0.01388	0.00154	0.36977	0.04152	0.31520	38.64827	0.04847	1.02293576	0.378249806	0.275988659
RUNOFF HMP 25 10 um	0.0524	274	17	1000	0.00325	0.00081	0.08787	0.02196	0.31520	27.64915	0.06775	1.0240963	0.08998462	-0.012276528
RUNOFF HMP 10-2 um	0.0524	261	18	1000	0.00361	0.00088	0.09767	0.02380	0.31520	26.33733	0.07112	1.03226844	0.100821645	-0.001439503
RUNOFF HMP 2-0 45 um	0.0524	359	173	1000	0.02523	0.00234	0.68247	0.06316	0.31520	36.22644	0.05171	1.02316393	0.698274217	0.596013069
RUNOFF HMP 0 45 um-10K De	0.0524	217	41	1000	0.00990	0.00169	0.26758	0.04557	0.31520	21.89732	0.08555	1.02709104	0.274828769	0.172587621
PROCEDURE BLANK	0.0524	363	27	1000	0.00369	0.00074	0.09984	0.01988	0.31520	36.64827	0.04847	1.0242749	0.102261148	0